Development of a new CO₂ injection process via Kenics-type static mixer for the ocean disposal of liquid CO₂

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ABSTRACT

A new type of the liquid CO₂ injection process via Kenics-type static mixer was proposed for the direct ocean disposal at the intermediate depths (500~1500 m). The flow of liquid CO₂ in a pipeline will be merged with the flow of pressurized water in the static mixer. In the mixer, the liquid CO₂ phase will be broken up into small drops in the water phase, and then the drops dispersed in the water flow will be discharged into the seawater. The performances of a static mixer have been investigated in a laboratory-scale experimental apparatus simulating the intermediate ocean conditions (500~700 m depth). The average drop size of liquid CO₂ was dramatically reduced with the use of the static mixer. In addition, the size distribution of liquid CO₂ drops after going through the static mixer was more uniform than that without the static mixer. The relationship between the average size of liquid CO₂ drops and the flow conditions of liquid CO₂ and water (Weber number) was experimentally determined. Under proper conditions, the flow of CO₂ hydrate-dispersed water was observed at the outlet of the static mixer without blockage of the pipe. Since the initial size and its distribution of the discharged liquid CO₂ drops would primarily determine the fate of the liquid CO₂ in the ocean, and consequently the environmental impacts, these results suggest that the use of the static mixer in the injection process has advantage in terms of controlling the environmental impact caused by the CO₂ disposal.

INTRODUCTION

Several scenarios of CO_2 ocean disposal have been proposed for a long-term sequestration. In the disposal processes, CO_2 captured and separated from the fuel gas of concentrated emission sources would be transported to the ocean through a pipeline, and then the CO_2 would be released to the seawater. The CO_2 would eventually dissolve in the seawater, and be sequestered for a certain period. The environmental impact by the dissolved CO_2 would depend on the various factors such as the releasing state of CO_2 (gas, liquid, solid), the releasing rate, size, and depth (shallow, <500m; intermediate, 500-1500m; deep, >3000m). In intermediate ocean disposal, especially, the CO_2 is released as the drops in the ocean. It is difficult to control the CO_2 drop size. The liquid CO_2 drops ascend into seawater because its weight is smaller than that of seawater. A proper design of the disposal process and an accurate estimation of the behavior of CO_2 in the ocean would be necessary for minimization of the environmental impact.

Then, we propose a new injection process of the CO_2 ocean disposal. **Figure 1** illustrates the outline of our process. A point of the process is the install of the static mixer. The liquid CO_2 pipeline is submerged into a depth of 500 to 1000m. In this depth, CO_2 is liquid and is also exchanged to CO_2

clathrate hydrate. A static mixer is installed into the end of pipeline. The mixer makes the injected CO₂ the formation of uniform-size drops and hydrate dispersed in seawater. The released CO₂ drops ascend or the hydrate particles descend with the weight in the ocean.

Figure 2 shows the schematic of the Kenics-type static mixer used in our experiments. Static mixer is referable to an in-line type motionless mixer, and is a simple and energy-saving mixing device. For the static mixer, mixing elements are arranged and fixed inside a straight pipe. For the Kenics-type static mixer, the element has a structure twisted the rectangular board 180° alternately with right and left. Then the element could make the incoming fluid to be turbulent. The flow in the static mixer is close to a piston flow, and uniform mixing could be realized. The energy consumption only derives from pressure drop in the static mixer. Then the power consumption is much lower than that by a stirred-tank type mixer. The mixers have been used in process requiring blending, reaction, dispersion, heat transfer and mass transfer [1].

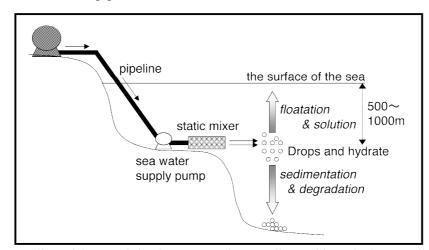


Figure 1. Outline of the new injection process in CO₂ disposal into the intermediate ocean

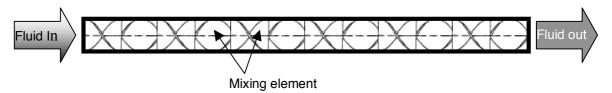


Figure 2. Schematic of the Kenics-type static mixer

By using the static mixer had those characteristics, the new injection process has several advantages:

1) Simplification of released CO₂ concentration control; this is because the CO₂ concentration of the release can be adjusted by regulating the mixing rate. This is effective in preventing a local concentration increase. 2) Simplification of droplet size control; the static mixer possesses this function.

3) Reduction of mass transfer rate into seawater; this is because the obtaining CO₂ drops have clathrate hydrate on the surface. The dissolution rate of hydrate into water is about one tenth times that of liquid CO₂. 4) Prevention of CO₂ pipeline blockage; under the CO₂ disposal condition, clathrate hydrate can be formed. The hydrate formation is a cause of the pipeline blockage. Since the static mixer disturbs a flow, blockage of the pipe by the hydrate is controlled. 5) Reduction of agitation energy; the energy consumption is only the energy based on the pressure drop in the mixer. 6) Simplicity of installation to equipment; it is only connecting the mixer to the end of the pipeline.

In this paper, we will discuss the laboratory simulation for this new process. We focused on the two points. They are, confirmation of CO₂ drops formation and investigation of effects of flows on drops sizes.

EXPERIMENTAL

Figure 3 shows the schematic of the experimental system for drops and hydrate formation. In the experiment, liquid CO2 from cylinder and water from tank were introduced into a Kenics-type static mixer. The purity of the liquid CO₂ was > 99.9% (supplied by Showa Tansan Co., Ltd) and the water was deionized and pre-cooled to a desired temperature. The flow rate of the high-pressure CO₂ pump (product of Nippon Seimitsu Co., Ltd., NP-AX-70) could be adjusted in a range from 23.2 to 93.4 ml/min and the flow rate of the high-pressure water pump (product of Fuji Pump Co., Ltd., 2JN224-10V) could vary from 472 to 2981 ml/min. The injection point cell is with parallel glass windows, diameter 25mm) due to the observation. The static mixer was designed and made especially for the experiment (by Noritake Co., Ltd, based on their product of type 3/8-N10-522N). One of the mixers was 210mm long and 10.9mm in inner diameter, and it had total of 12 mixing elements with a lengthto-diameter ratio of 1.5; the mixer was made of SUS316. One or two mixers were installed in the apparatus. A transparent polycarbonate section, with 10.6mm in inner diameter and 300mm in length, was in the downstream of the mixer for observing the release from the mixer. The temperature of the CO₂-water mixture was controlled by the cooling jacket, through which the coolant (a mixture of water and ethylene glycol) from a thermal bath was circulated. The temperature of the system could be controlled to ± 0.1 K. The backpressure valve controls the pressure in the system. At any set value, fluctuations in the system pressure agreed within 0.01 MPa. The pressure durability of the apparatus is not more than 10 MPa.

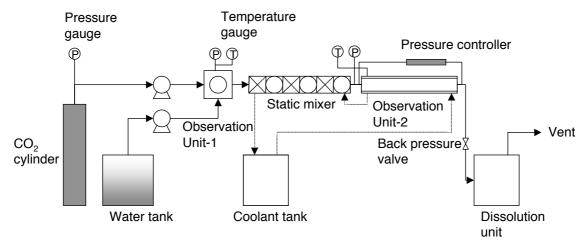


Figure 3. Schematic drawing of the experimental apparatus

The typical experimental pressure and temperature were, respectively, 7.0 MPa and 277 K. The typical flows for liquid CO₂ and water were 46.6 ml/min and 2.06 L/min, respectively. In the experiment, the two-phase pattern of the mixture released from the mixer could be observed in the observation section, and it was recorded by the high-speed video camera (FOR.A Co. Ltd., VFC-1000 and Photron Ltd., FASTCAM-Net500). The captured video data then were analyzed in a Power Macintosh G4 computer using the ImageJ-1.27 program developed by the U.S. National Institute of Health.

RESULTS AND DISCUSSION

Agitation effect of the static mixer on the CO₂-water flow

Figure 4 shows a typical observed results. In Fig.4, the (a) shows CO_2 -water mixture after through empty pipe, and the (b) shows the released mixture from the static mixer. In both cases, the released mixture was a disperse- CO_2 drop flow, and the CO_2 drops had a hydrate phase on the surface. The

disperse- CO_2 drop flow as (b) was always obtained by the control of water flow rate. Such condition was when the Reynolds number of the continuous flow based on the inner diameter of the mixer, Re > 2300, that is turbulent flow. When Re < 2300 (laminar flow), the CO_2 drops agglomerated by hydrogen bond between hydrates at the surface, then the released mixture was the plug-like flow.

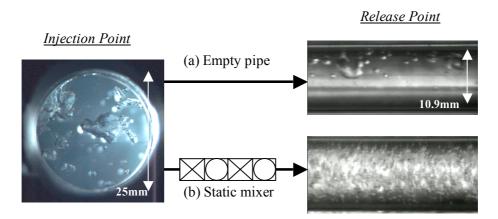


Figure 4. Observation results of CO₂-water flow

The average diameter of the drops can be explained by the Sauter mean diameter (SMD) as follows equation;

$$SMD = \prod_{i} N_i D_i^3 / \prod_{i} N_i D_i^2.$$
 (1)

Here, N_i is the number of drops for the *i*th group having a diameter D_i . Under the condition shown in Fig. 4, SMD = 2.64 mm at the injection point, 1.63 mm for empty pipe (a) and 0.40 mm for static mixer (b) at the release point. The SMD for the release point was smaller than that of the injection point, and then, in the cases of various water flow rate, SMD for static mixer was smaller than that of empty pipe. The results indicate that the drops is more broken up by the shear stress in the empty pipe and the static mixer, especially, the drops are agitated and broken up in the static mixer. **Figure 5** shows the cumulative frequency of drop diameter of the observation results in Fig. 4. For the static mixer agitation, the drop diameter varies from 0.2 to 0.6 mm, and the variance of drop diameter $\Box_D^2 = 0.00114$. On the other hand, the range of the drop diameter for the empty pipe is from 0.3 to 2.5 mm, and $\Box_D^2 = 0.110$. In the CO₂ injection point, the diameter range is to 6.2 mm, and $\Box_D^2 = 2.103$. The standard deviation of the drop diameter \Box_D for the static mixer is one tenth more than that for the empty pipe. Fig. 5 is clear that the agitation by the static mixer makes CO₂ drops small and uniform in size under high pressure and low temperature condition, therefore, the injection process will make the environmental impact by the CO₂ ocean disposal reduce.

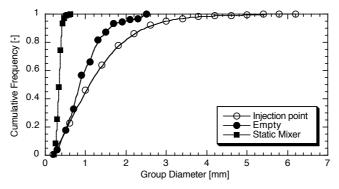


Figure 5. Typical cumulative frequency of CO₂ drop diameter

Influence of continuous phase flow on the drop size

Drop formation depends mainly on forces resulting in surface tension and fluid dynamics forces. Then, *SMD* is relation with the Weber number, *We*. The *We* is defined as

$$We = \frac{D \square u^2}{\square}, \qquad (2)$$

where \square presents the density of the fluid, \square the interfacial tension. In the case of the static mixer, D is the inner diameter of the mixer (D_{mixer}), \square the density of the continuous-phase fluid, and u the linear velocity of the fluid [2]. Assuming that CO_2 hydrate, which thickness is negligible in comparison with the drop diameter, is formed on the surface of liquid CO_2 drop, the hydrate contacts water. Because the maximum of the mole fraction of liquid CO_2 in the experimental conditions is 0.03, the effect of liquid CO_2 on the density of the continuous flow is negligible.

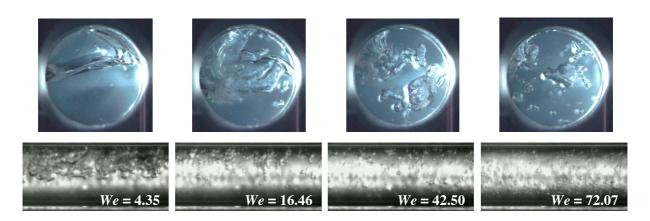


Figure 6. Observation results in the various Weber number Upper: injection point, lower; release point from the static mixer

Figure 6 shows observation results of the CO_2 -water mixture flow by changes in the Weber number which varies with the water flow rate. At We = 4.35 and 16.46, in which the flows are the laminar flow condition, the plug-like flow was observed. The turbulent flow condition is necessary to gain the dispersed- CO_2 flow.

Based on above conditions, SMD of the observation data are plotted as a function of We in **Figure 7**. The Weber number and a function of static mixer are very dominant in SMD. SMD can be expressed with a function of We like $SMD/D_{mixer} = AWe^B$ in both cases. Thus, Weber number governs the size of liquid CO_2 drops. With respect to drop size, the maximum continuous flow rate kept the agitation effect by the Kenics-type static mixer in this apparatus can be estimated from those equations. This point is where the two line meet, this is We = 2840. When this Weber number, the flow rate is calculated to be about 41.9 L/min.

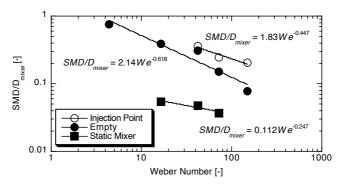


Figure 7. Relationship of *SMD* with *We* number

Influence of dispersed phase flow on the drop size

Figure 8 plots the SMD at the release point as a function of the liquid CO_2 flow rate. The slope of the line decreases gradually with Reynolds number. When turbulent flow region, the change of the slope will almost be lost, and the relationship of SMD and liquid CO_2 flow rate is very poor in the range of this experiment. This tendency depends on the initial size of the drops injected into the static mixer. Since the cross-section area of the liquid CO_2 nozzle is fixed in Fig. 8, the linear velocity of CO_2 increases with the increase in liquid CO_2 flow rate. The increase in the velocity makes the break-up of the liquid CO_2 fluid easy, and the initial size of the CO_2 drops small. When turbulent flow condition, since the water flow rate effect on the initial drop size is larger than the CO_2 flow rate effect, the CO_2 flow rate effect seems to be poor.

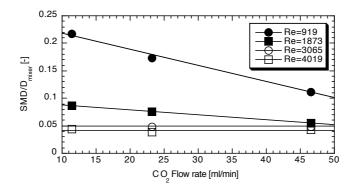


Figure 8. Effect of liquid CO_2 flow rate on the drop diameter *SMD*

*Influence of the agitation time on the CO*₂-water flow

Figure 9 shows the effect of the change in the agitation time by the static mixer on the drop size. Agitation by the static mixer can give the uniform size drops in a very short agitation time. The agitation time was controlled by the change in the number of the mixing element. Since the flow in the static mixer is similar to a piston flow, the agitation time Γ is estimated by the following equation,

$$\Box = \frac{V_{mixer}}{Q_{total}} \ . \tag{3}$$

Here, Q_{total} is the total flow rate of liquid CO_2 and water, and V_{mixer} is the inner volume of the static mixer. When the empty pipe, $\Box = 0$. The SMD decreases dramatically by the agitation. Since it is known that the micro-mixed environment of the first two elements of Kenics-type static mixer, $\Box = 0.2$ under the condition in Fig. 9, is better than the subsequent elements of the mixer [3], the SMD may decrease dramatically at the first two elements, the agitation time between 0 and 0.2s. At the subsequent elements, the SMD decreases gradually.

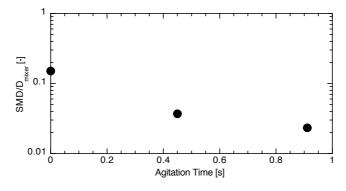


Figure 9. Effect of the agitation time by the static mixer on the drop size

The increase in the agitation time gave us another interesting observation result. That is the hydratewater flow. Figure 10 shows the typical observation results. As shown in Fig. 10, the hydrate plug-like flow was observed at the release point. In addition, this plug-like hydrate did not cause the pipeline blockage. If this hydrate is distributed, it will become hydrate dispersed flow. Moreover, a hydrate particle may be able to be formed if these hydrates are coalesced. Since the hydrate dispersed flow is lower CO₂ concentration than drop dispersed flow, release of the hydrate dispersed flow to the ocean will prevent the increase in an environmental impact.

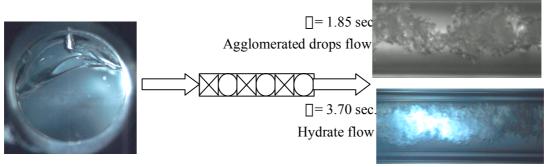


Figure 10. Hydrate flow generation by the change in the agitation time Condition: CO₂ flow 93.4 ml/min, Water flow 0.47 L/min

CONCLUSION

A new type of the liquid CO₂ injection process via Kenics-type static mixer was proposed for the direct ocean disposal at the intermediate depths (500~1500 m). The performances of a static mixer were investigated in a laboratory-scale experimental apparatus simulating the intermediate ocean conditions (500~700 m depth). The average drop size of liquid CO₂, which was represented as the Sauter mean diameter (SMD), was dramatically reduced with the use of the static mixer. In addition, the size distribution of liquid CO₂ drops after going through the static mixer was more uniform than that without the static mixer. The water flow rate, liquid CO₂ flow rate, that is Weber number (We), and agitation time were influence on the SMD of the liquid CO2 drops. The relationship between the average size of liquid CO2 drops and the flow conditions of liquid CO2 and water, was experimentally determined. The SMD could be plotted as a function of We, $SMD/D_{mixer} = AWe^{B}$, where A and B is constant. Under proper conditions, the flow of CO2 hydrate-dispersed flow was observed at the outlet of the static mixer without blockage of the pipe. Since the initial size and its distribution of the discharged liquid CO₂ drops would primarily determine the fate of the liquid CO₂ in the ocean, and consequently the environmental impacts, these results suggest that the use of the static mixer in the injection process has advantage in terms of controlling the environmental impact caused by the CO₂ disposal.

NOTATION

A: constant B: constant D: drop diameter

mixer inner diameter D_{mixer} : N: number of drop

total flow rate Q_{total} : Reynolds number Re: Sauter mean diameter SMD:

u: linear velocity of the fluid

 V_{mixer} : inner volume of the static mixer

We: Weber number

 \Box : density of the fluid

☐: interfacial tension

 \square_D : standard deviation of the drop diameter

 \square_D^2 : variance of the drop diameter

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